Chemosphere 199 (2018) 44–53

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Sorption and bioaccumulation behavior of multi-class hydrophobic organic contaminants in a tropical marine food web

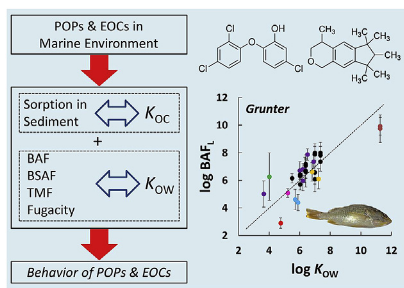
Hui Zhang, Barry C. Kelly*

Department of Civil and Environmental Engineering, National University of Singapore, Singapore

HIGHLIGHTS

- 90 HOCs were analyzed in tropical marine food web.
- Strong linear relationship between K_{OC} and C_{SED}/C_{WD} was observed.
- BAF_L, BSAF, fugacity ratio and TMF were applied for comprehensive evaluation.
- Galaxolide and methyl triclosan had relatively high bioaccumulation potential.
- Triclosan, tonalide, DPs and other novel BFRs had low bioaccumulation potential.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 23 September 2017
Received in revised form
31 January 2018
Accepted 31 January 2018
Available online 1 February 2018

Handling Editor: Myrto Petreas

Keywords:
Hydrophobic organic contaminants
Sorption
Bioaccumulation
Tropical
Marine

ABSTRACT

While numerous studies have demonstrated the environmental behavior of legacy persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), information regarding sorption and bioaccumulation potential of other widely used organic chemicals such as halogenated flame retardants (HFRs) is limited. This study involved a comprehensive field investigation of multi-class hydrophobic organic contaminants (HOCs) in environmental media and fish in Singapore Strait, an important tropical maritime strait in Southeast Asia. In total, 90 HOCs were analyzed, including HFRs, synthetic musks, PCBs, OCPs, as well as triclosan and methyl triclosan. The results show that the organic carbon normalized sediment-seawater distribution ratios (C_{SED}/C_{WD}) of the studied compounds are comparable to the organic carbon-water partition coefficients (K_{OC}), over a log K_{OC} range of approximately 4–11. The observed species-specific bioaccumulation factors (BAFs), biota-sediment accumulation factors (BSAFs), organism-environment media fugacity ratios (f_{FISH}/f_{WD} and f_{FISH}/f_{SED}) and trophic magnification factors (TMFs) indicate that legacy POPs and PBDE 47 show bioaccumulation behavior in this tropical marine ecosystem, while triclosan, tonalide, dodecachlorodimethanodibenzocyclooctane stereoisomers (DDC-COs), and hexabromocyclododecanes (HBCDDs) do not. Methyl triclosan and galaxolide exhibit moderate biomagnification. Tetrabromobisphenol A (TBBPA) and 1,2-bis (2,4,6-tribromophenoxy)ethane (BTBPE) were detected in environmental media but not in any of the organisms, suggesting low bioaccumulation potential of these flame retardants. The apparently low bioaccumulation potential of the studied HFRs and synthetic musks is likely because of

* Corresponding author. Department of Civil and Environmental Engineering, National University of Singapore, Block E1A, #07-03, No.1 Engineering Drive 2, 117576, Singapore.
E-mail address: bc_kelly@sfu.ca (B.C. Kelly).

https://doi.org/10.1016/j.chemosphere.2018.01.173

metabolic transformation and/or reduced bioavailability due to the hydrophobic nature of these compounds.

1. Introduction

Occurrence of triclosan, methyl triclosan, synthetic musks (e.g. galaxolide and tonalide), chlorinated flame retardants (CFRs, e.g. dechlorane plus isomers or dodecachlorodimethanodibenzocyclooctane isomers: *syn* and *anti*-DDC-COs) and brominated flame retardants (BFRs, e.g. hexabromocyclododecane isomers: α, β and γ- HBCDDs) in environment has been recorded in recent years (de Wit, 2002; Heberer, 2002; Hale et al., 2003; Covaci et al., 2006; Law et al., 2006b; Wang et al., 2007; Brausch and Rand, 2011; Covaci et al., 2011; Sverko et al., 2011; Bedoux et al., 2012; Liu and Wong, 2013). The bioaccumulation behavior of these hydrophobic organic contaminants (HOCs) in various environmental ecosystems has been attracting much attention worldwide, due to their wide distribution, possible persistent properties and toxic effects (Eljarrat and Barcelo, 2003; Brausch and Rand, 2011; Marvin et al., 2011; Bedoux et al., 2012; Costa et al., 2014). To better understand the bioaccumulation behavior of contaminants, many field estimated factors have been recommended and applied, including bioaccumulation factor (BAF), biota-sediment bioaccumulation factor (BSAF), biomagnification factor (BMF) and trophic magnification factor (TMF) (Arnot and Gobas, 2006; Borga et al., 2012b; Conder et al., 2012; Mackay et al., 2013). However, the comprehensive investigation of bioaccumulation behavior of above-mentioned HOCs through the field estimated factors is still limited until now.

It is well-known that synthetic musks, triclosan and methyl triclosan are ubiquitous in environment, which is found to be tightly connected to the discharge of wastewater treatment plants (Lindstrom et al., 2002; Singer et al., 2002; Buerge et al., 2003; Bester, 2005; Horii et al., 2007). There are only a few studies that investigated the bioaccumulation behavior of synthetic musks, and these studies provided non-consistent results. For example, Nakata et al. (2007) reported the possible trophic dilution of galaxolide by showing the negative correlation between galaxolide concentration and the trophic levels of organisms in marine food web, even though the TMF was not estimated (Nakata et al., 2007). However, Zhang et al. (2013) showed that galaxolide might slightly biomagnified in Taihu Lake food web (TMF: 1.12, $p > 0.05$) (Zhang et al., 2013). To the best of our knowledge, the bioaccumulation behavior of triclosan and methyl triclosan has not yet been thoroughly evaluated in any food web yet.

As typical flame retardants, polybrominated diphenyl ethers (PBDEs) have been well studied for their bioaccumulation behavior in marine environment, lakes and reservoirs, showing the predominant biomagnification of PBDE 47 among the congeners (Law et al., 2006a; Kelly et al., 2008; Tomy et al., 2008a; Wan et al., 2008; Wu et al., 2008; Debruyn et al., 2009; Hu et al., 2010). But there are only limited studies for non-PBDE BFRs, such as HBCDDs, hexabromobenzene (HBB), pentabromoethylbenzene (PBEB), pentabromotoluene (PBT), tetrabromobisphenol A (TBBPA), and 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE). Studies found that α-HBCDD was preferentially enriched in food web compared to β & γ-HBCDDs, mainly due to their different metabolism rates in biota body (Zegers et al., 2005; Covaci et al., 2006; Tomy et al., 2008a, 2011; Wu et al., 2010a). Studies also found that TBBPA, BTBPE, PBT and PBEB all had low bioaccumulation potential in marine or freshwater food webs (Morris et al., 2004; Law et al., 2006a; Wu et al., 2010a), but HBB experienced trophic magnification in one freshwater food web near e-waste recycling site in China (Wu et al., 2010a).

DDC-CO isomers are found to behave differently in different food webs. For examples, DDC-COs experienced trophic dilution in the Lake Ontario food web (TMFs: 0.44 and 0.34) (Tomy et al., 2007), but they showed trophic magnification behavior in other freshwater and marine food webs (TMFs: 1.6–11.3) (Tomy et al., 2007; Wu et al., 2010b; Peng et al., 2014; Sun et al., 2015). Also, previous studies found that the ratio of *syn*-DDC-CO to *anti*-DDC-CO was lower in biota than sediment or water (Tomy et al., 2007; Wu et al., 2010b; Jia et al., 2011), and one study even found that the ratio kept decreasing along with the increase of trophic levels (Wu et al., 2010b). However, another study showed that the ratio of *syn*-DDC-CO to *anti*-DDC-CO was higher in biota than abiotic samples in the Lake Winnipeg food web (Tomy et al., 2007).

The previous studies of occurrence and bioaccumulation behavior of HOCs focused on freshwater and marine food webs in arctic and temperate areas, and either BAF/BSAF or TMF was used as the key indicator. The comprehensive evaluation of bioaccumulation behavior of HOCs in food webs through BAF, BSAF and TMF is still rare. Especially, not only the emerging organic contaminants (EOCs, e.g. synthetic musks, triclosan, methyl triclosan and flame retardants), but also the legacy persistent organic pollutants (POPs), such as polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs), have not been well studied for their occurrence and bioaccumulation behavior in tropical marine food web so far. Located near the equator in Southeast Asia, Singapore Strait provides an ideal site of tropical marine environment greatly influenced by human activities. In this study, various samples, including seawater, marine sediment, suspended particulate organic matter (SPOM) and fish, were collected in Singapore Strait and analyzed for EOCs and legacy POPs. The major objective was to provide comprehensive evaluation of bioaccumulation behavior of HOCs in tropical marine environment through BAF, BSAF and TMF. The fugacities of HOCs in Singapore Strait food web and the sorption behavior of HOCs between marine sediment and seawater were studied to facilitate the understanding of the bioaccumulation behavior of HOCs.

2. Materials and methods

2.1. Chemicals and materials

In this study, a total of 90 HOCs were analyzed, including 28 PCBs, 22 OCPs, 7 chlorobenzenes, 10 synthetic musks, 19 BFRs, 2 CFRs, as well as triclosan and methyl triclosan. Stable-isotope, carbon-13 ($^{13}C_x$) and deuterium (d_x), labelled compounds were used as internal surrogate standards (IS) or injection recovery standards (RS). The detailed information of the target analytes, organic solvents and other materials is provided in the Supporting Information (SI, Table S1).

2.2. Sample collection

Between 2011 and 2012, samples of seawater, marine sediment,